

Zeolite-filled PMMA composite membranes: influence of coupling agent addition on gas separation properties

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Abstract

The adhesion between the polymer phase and the external surface of the particle appeared to be a major problem in the preparation of zeolite-filled polymeric membranes when the polymer is in the glassy state at room temperature. Interfacial void-free PMMA membranes filled with zeolite 4A were prepared by introducing 3-(trimethoxysilyl) propylmethacrylate (TMOPMA) as a way to improve the gas separation properties of the polymeric membranes. It was found that the oxygen permeability of zeolite-filled PMMA membranes was lower than pure PMMA membranes. In addition, the oxygen permeability of zeolite-filled PMMA membranes decreased with increasing zeolite 4A loading up to 33.3 wt%. At the same zeolite composition, zeolite surface modification with TMOPMA significantly improved the solubility and diffusivity coefficients of the zeolite-filled PMMA membranes. The sorption behaviors of PMMA/zeolite 4A and PMMA/zeolite 4A/TMOPMA membranes can be well described by the dual-mode sorption model. Surface modification of the zeolite particles increases the gas sorption in the Henry sites but decreases the gas sorption in the Langmuir sites. The solubility of the zeolite-filled PMMA membrane is predicted by the rule of mixtures at a zeolite loading less than 20 wt%.

Keywords: Zeolite-filled PMMA membranes; Gas separation; Dual-mode sorption

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1. Introduction

The development of polymeric membranes for the separation and purification of gases based on the selective permeation of one component of a mixture has attracted a great deal of interest during the last decades [1–3]. Intensive investigation of gas separation characteristics of polymeric membranes has resulted in rapid improvements in their properties [4–7]. In the early 1990s, an upper limit of performance for polymeric membranes was noted for the separation of oxygen from nitrogen [8]. This limit of performance for polymeric membranes was below the desirable performance level for practical applications, while on the same scale carbon molecular sieve and zeolite membranes exhibited commercially attractive properties for the separation of oxygen from nitrogen [9–11]. Although molecular sieving materials lie well above the upper bound polymeric trade-off curve, these materials are expensive and difficult to process as membranes. Drawbacks in both the polymeric and purely molecular sieving materials suggest the need for a hybrid approach to membrane material development and membrane processing [12,13].

The effect of zeolite particles incorporated in a polymer on transport properties of membranes made from these polymers has received much attention recently [14–19]. Zeolite-filled rubbery polymer membranes were studied by Jia et al. [20]. They concluded from their results that silicalite-filled PDMS membranes have higher O_2 permeabilities and better O_2/N_2 selectivities than unfilled membranes. Silicalite plays the role of a molecule sieve in the silicalite-filled membranes. Suer et al. [14] investigated the gas permeation characteristics of polyethersulfone–zeolite mixed matrix membranes. His conclusion was that permeabilities first decrease and then increase with increasing zeolite loading. At high zeolite loadings (42–50 wt%), both permeabilities and selectivities increase. Changes in membrane performance were not only due to zeolite crystals,

but also depended on the complex heterogeneous micromorphology, including the lack or presence of zeolite particle aggregation, and the cave-like voids created around the zeolitic filler. Mahajan et al. [21] incorporated 4A zeolite into polyimide. They found that two factors seem to be critical to the formation of the nonselective defects at the interface: the nature of the polymer-sieve interaction, and the stress encountered during material preparation. Duval et al. [22] found that permeabilities increased but selectivities decreased or maintained when silicalite was added into glassy polymers. They attributed this result to the formation of interfacial voids due to the poor adhesion of the glassy polymer and the zeolite surface. Zeolite particles treated with a silane coupling agent resulted in considerable improvement of the internal structure of silicalite-filled PEI membranes. However, permeation results could not illustrate this improvement.

The results mentioned above have never shown any clear explanation why zeolite surface modification could change the gas transport properties of zeolite-filled glassy polymer membranes. In this work, 3-(trimethoxysilyl)propylmethacrylate (TMOPMA) was introduced as a kind of compatibilizer to eliminate the interfacial void. Pure gas permeation and sorption of surface modification zeolite-filled PMMA membranes were investigated to find the reason why zeolite surface modification affects the zeolite-filled glassy polymer membrane performance.

2. Experimental

Poly(methyl methacrylate) (MW: 996,000, Aldrich) was chosen as the matrix material for the composite membrane. Zeolite 4A (Aldrich) was chosen as the filler. TMOPMA (Aldrich) and tetrahydrofuran (THF, Tedia) were used as received.

The TMOPMA is mixed with distilled water and then the zeolite is added. The reacting

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